High Pressure-High Temperature Synthesis and Elasticity of the Cubic Nitride Spinel γ-Si_3N_4

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Introduction: The compressional behavior of a new dense form of silicon nitride with the cubic spinel structure is studied be energy dispersive X-ray diffraction, following *in situ* synthesis from the low pressure form by laser heating in the diamond anvil cell. The unit cell dimension and the ambient temperature bulk modulus and its pressure derivative are determined to be V_0 = 8.29(3) Å^3/atom, K_0 = 308(5) GPa, for K'_0 = 4, in excellent agreement with theoretical calculations.

Dense nitride structures of Group III and Group IV elements (Al, Si, Ga) are well known technological materials, with high mechanical strength and hardness and useful semiconducting properties[1,2]. Recently new forms of nitrides Si_3N_4 and Ge_3N_4 were found following high-pressure high-temperature synthesis.

Methods and Materials: In this study, we have obtained the cubic γ-spinel phase of Si_3N_4 by laser heating low pressure (α- and β-) forms of silicon nitride in the DAC, and have studied the new phase and its decompression behavior by synchrotron (energy dispersive) X-ray diffraction. The results permit the determination of the zero-pressure bulk modulus and cell volume. Commercially obtained α-Si_3N_4 (containing ~5-10% β-Si_3N_4) was loaded into a symmetric diamond cell for double-sided laser heating studies. Samples were loaded with ~1% amorphous boron powder to efficiently absorb the heating laser energy, and were pressurized to approximately 20 GPa. Laser heating was achieved with a Nd:YAG laser. Energy dispersive X-ray diffraction patterns were collected at the superconducting wiggler beam line station X17B1.

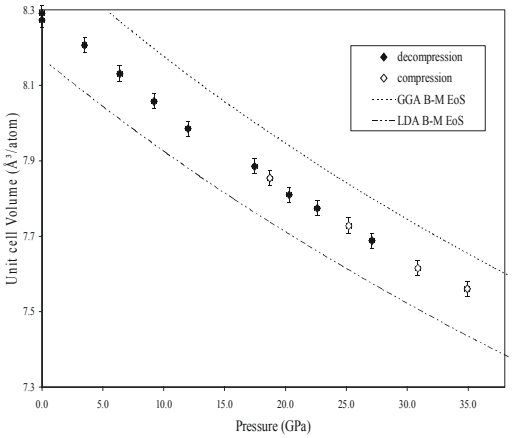

Figure 1. Pressure dependence of the unit cell volume in Si_3N_4 spinel measured between ambient and 35 GPa at room temperature. The open symbols are from the data collected during compression and the filled symbols are from the data collected during decompression. The dotted line is the GGA result while the dash-dotted line is the LDA result.

Results: The diffraction patterns collected at room pressure (after complete decompression) gave the value of V_0 = 8.29(3) Å^3/atom. The calculated values of V_o and K_o obtained from DFT calculations compare well with the experimental values. The LDA underestimates the V_o by 1,7 % but agrees with the bulk modulus within experimental error. The GGA overestimates V_o by about 2.0% and underestimates K_o by about 7.8 %.

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References: [1]A.Y. Liu and M.L. Cohen,"Structural properties and electronic structure of low-compressibility materials: β-Si_3N_4 and hypothetical β-C_3N_4" **41**, *10727*, Phys. Rev. B 1990. [2] N.E. Christensen and I. Gorczyca, "Optical and structural properties of III-V nitrides under pressure" **50**, *4397*, Phys. Rev. B, 1994